—研究論文—
Scientific Paper

Evaluation of SO_2 Emission from the 1982 Eruption of El Chichon by Glaciological and Satellite Methods

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南極の積雪試料を用いたエル・チチョン火山1982年噴火に伴う二酸化硫黄放出量の推定

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要旨: 火山噴火に伴う大気中への硫黄放出量の推定方法の一つに，極地方の氷床の硫酸堆積量に基づく方法がある．これは，氷床に堆積した雪に含まれる火山性硫酸の量にスケーリングファクター（核実験により放出され成層圏を経て降下した放射性物質の広がり方に基づいて決められた，成層圏を経由する物質運搬効率を地球上の任意の地点で与えるファクター）を乗じる方法である．ケーススタディとして，近年の爆発的噴火の一つであるメキシコのエルチチョン火山1982年噴火について，南極氷床のドームふじ雪試料から硫黄放出量を推定した．同噴火による放出量は，人工衛星に搭載した紫外線分光器（TOMS）による噴煙のリモートセンシングにより推定されており（7 Mt SO_2），本研究による推定値はこれの2.5倍程度の値であった（17 Mt SO_2）．このことは，過去の大噴火に伴う硫黄放出量の推定に，氷床の硫酸堆積量に基づく方法が適用できることを示している．

Abstract: The SO_2 emission at the time of the 1982 eruption of El Chichon in Mexico was estimated by a glaciological method based on sulfate ion concentration and bomb-produced total β activity in snow collected in Antarctica. The glaciological estimate of the SO_2 emission is 17 ± 1 Mt SO_2, somewhat greater than the spectroscopic estimate (7 Mt SO_2) measured by a satellite-carried total ozone mapping system. We conclude that the glaciological method is useful to estimate SO_2 emissions of past explosive volcanic eruptions in the equatorial region.

1. Introduction

Major explosive volcanic eruptions emit a large amount of sulfur dioxide (SO_2) into the stratosphere. The SO_2 is gradually converted into sulfuric acid aerosols in the stratosphere, spreads over the globe, and eventually falls onto the Earth's surface in a few years. Snow that accumulated continuously on polar ice sheets contains a record of such volcanic sulfate signals. The stratospheric sulfuric acid aerosols reduce atmospheric transmissivity and temperature at the Earth's surface. The effects on climate of

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recent explosive eruptions such as Agung in 1963, St. Helens in 1980, El Chichon in 1982, and Pinatubo in 1991 have been well-documented (DEVINE *et al.*, 1984; RAMPINO and SELF, 1984; MINNIS *et al.*, 1993; SELF *et al.*, 1996). For evaluation of the relationship between volcanism in the past and subsequent climatic change, it is essential to estimate the amount of SO_2 emitted by past explosive volcanic eruptions.

Volcanic SO_2 emission has been estimated by (1) the glaciological method, (2) the petrological method, and (3) the spectroscopic method. The glaciological method is based on the SO_4^{2-} ion concentration in a polar snow sample that accumulated at the time of a volcanic eruption, multiplied by the local accumulation rate of snow and the ratio of locally measured bomb-produced total β activity to the total β activity produced at the time of atmospheric bomb tests in the equatorial region in 1952 and 1954 (HAMMER *et al.*, 1980; CLAUSEN and HAMMER, 1988; LANGWAY *et al.*, 1988; ZIELINSKI, 1995). The petrological method is based on the difference of sulfur concentrations between glass inclusions in phenocrysts and matrix glasses scaled by the mass of the magma erupted. Glass inclusions are considered to preserve pre-eruptive dissolved volatile concentration in melt of the magma, whereas matrix glasses have lost the dissolved volatiles during decompression and eruption of the magma (SIGURDSSON, 1982; DEVINE *et al.*, 1984; PALAIS and SIGURDSSON, 1989; KOHNO, 1992; MANDEVILLE *et al.*, 1996). The spectroscopic method, which has been established as a direct method to measure SO_2 emission, is based on UV spectrometric measurement in the volcanic cloud by a satellite-carried total ozone mapping system (TOMS: KRUEGER, 1983; KRUEGER *et al.*, 1990; BLUTH *et al.*, 1992).

Recently it has been pointed out, particularly since spectroscopic measurements have become available, that the petrological estimates are generally much lower than the spectroscopic estimates by a factor of 10 or more, the spectroscopic method being regarded as providing plausible estimates of SO_2 emission (ANDRES *et al.*, 1991; WESTRICH and GERLACH, 1992; GERLACH *et al.*, 1994; KAZAHAYA and SHINOHARA, 1994). The difference is referred to as excess sulfur degassing. The cause of excess sulfur degassing is currently one of the most active research topics in volcanology (DEVINE *et al.*, 1984; RAMPINO and SELF, 1984; KRUEGER *et al.*, 1990; ANDRES *et al.*, 1991; WALLACE and GERLACH, 1994; GERLACH *et al.*, 1994; HATTORI, 1996; RUTHERFORD and DEVINE, 1996; KRESS, 1997). Because of their own nature, the glaciological and petrologic methods are only used to estimate SO_2 emission by historic volcanic eruptions.

In this study, we compare the spectroscopic and glaciological estimates of volcanic SO_2 emission by the 1982 eruption of El Chichon in Mexico. The eruption was one of the most explosive in recent years. The amount of SO_2 emission estimated by the spectroscopic and petrologic methods have been reported to be 7 Mt SO_2 (BLUTH *et al.*, 1993) and 0.07 Mt SO_2 (DEVINE *et al.*, 1984), respectively, although the amount of the emission estimated by the glaciological method has not been reported yet. We estimated the SO_2 emission by the glaciological method using snow samples collected at Dome Fuji Station, Antarctica.

2. Samples and Analytical Procedures

Snow samples were collected in January 1997 from the wall of a 3.8 m deep pit

excavated at Dome Fuji Station (77° 19′01″S, 39° 42′12″E, 3810 m a.s.l.) located at the summit of the inland plateau in Queen Maud Land, East Antarctica (Fig. 1). Characteristic of snow stratification observed on the pit-wall was uniform bedding with well-developed depth hoar. Snow samples were collected from 57 layers between surface and 3.75 m depth and stored in 100 m*l* polyethylene bottles. The stratigraphy of the upper 36 snow layers is shown in Fig. 2a.

The annual average accumulation rate at Dome Fuji Station has been reported to be 32 $kg \cdot m^{-2} \cdot a^{-1}$ (KAMIYAMA *et al.*, 1989). This was estimated from the tritium peak of 1964/65 which appeared at 0.63 m in water equivalent depth of snow collected at DF-80, 2 km southwest from Dome Fuji Station, in December 1985 (KAMIYAMA *et al.*, 1989). Snow layers of the pit collected in January 1997 were dated using the annual accumulation rate as shown in Fig. 2.

The snow samples, preserved in a low-temperature room (−20° C), were melted in a refrigerator (+5° C) overnight. Concentrations of anions (Cl^-, F^-, SO_4^{2-}, NO_2^- and NO_3^-) and cations (Na^+, K^+, NH_4^+, Mg^{2+} and Ca^{2+}) were simultaneously measured using an ion chromatograph (Dionex model DX-500) equipped with AG11 and CG14 guard columns, AS11 and CS14 separator columns, and ASRS-1 and CSRS-1 auto suppressors to concentrate the ionic species having a concentration level of ~10 $ng \cdot g^{-1}$. The precision and detection limit of the measurement for each ion are

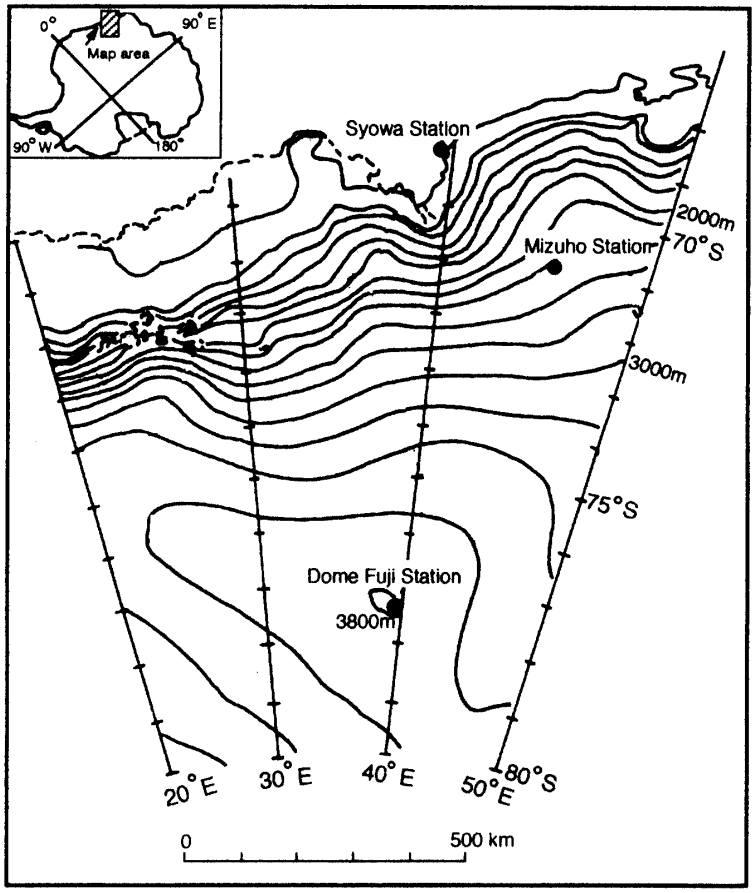

Fig. 1. Location of Dome Fuji Station in Queen Maud Land, East Antarctica.

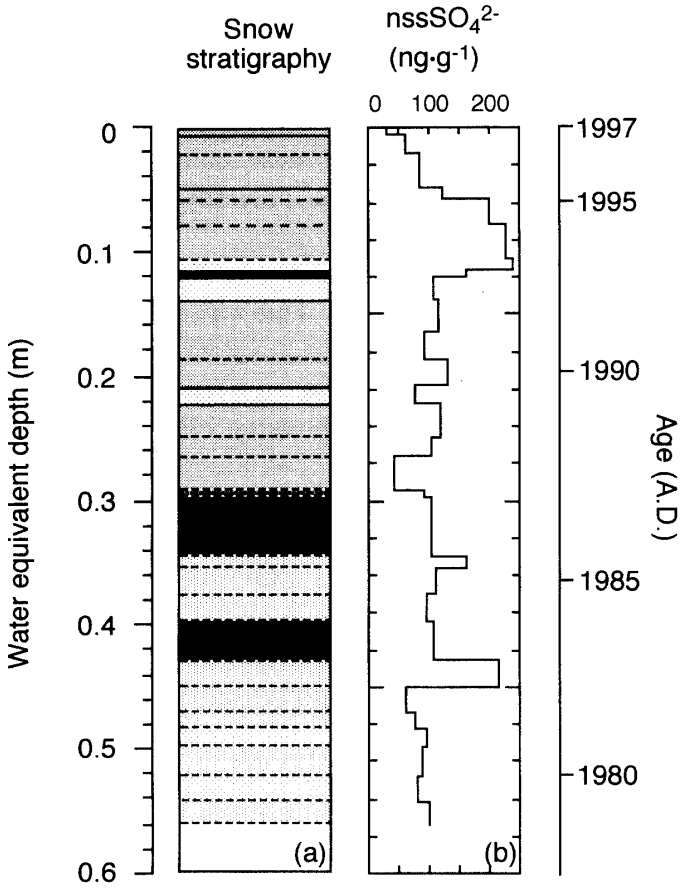

Fig. 2. (a) Stratigraphy and (b) non-sea-salt sulfate ($nssSO_4^{2-}$) concentration of the upper 36 snow layers of a 3.8 m deep snow pit excavated in January 1997 at Dome Fuji Station, East Antarctica. In the stratigraphic diagram, solid and dotted lines indicate layer boundaries with and without ice crust, respectively. Hatched shade indicates the degree of hardness (a harder layer is darker).

described in WATANABE *et al.* (1997).

3. Basic Principle of the Glaciological Method

The total amount of sulfur from a source volcano can be estimated under the following assumption. The ratio of the amount of sulfur reaching the snow pit site to sulfur emitted from a source volcano in the equatorial region is assumed to be equal to the ratio of total β activity observed at the pit site to the total β activity produced by atmospheric bomb tests in the same region (HAMMER, 1977; HAMMER *et al.*, 1980). This is expressed by the following equation:

$$SO_2 = [SO_4]^*_{snow} \cdot \frac{\beta^{\text{test region}}}{\beta^{\text{pit site}}} \cdot \frac{W_{SO_2}}{W_{H_2SO_4}}, \quad (1)$$

where $[SO_4]^*_{snow}$ is the amount of volcanic sulfate (*i.e.* non-sea-salt sulfate as described later) deposition in snow sample, $\beta^{\text{test region}}$ is the amount of total β activity produced by the atmospheric nuclear bomb tests and $\beta^{\text{pit site}}$ is the amount of total β activity measured

at the pit site. W_{SO_2} and $W_{H_2SO_4}$ denote the molecular weights of SO_2 and H_2SO_4, respectively.

The total β activity consists mainly of ^{90}Sr and ^{137}Cs (CLAUSEN and HAMMER, 1988). The basic concept behind this assumption is that the transport of aerosols is controlled by the global atmospheric circulation and that the behavior of sulfuric acid aerosols of volcanic origin and the bomb-produced aerosols are the same on a global scale.

The total β activity at the pit site also needs to be measured. According to the UNSCEAR Report (1982), the total β activity from the US atmospheric bomb tests at 11°N in 1952 and 1954 was 9.56 MCi. Then, sulfur (in terms of SO_2) emission by volcanic eruption in the equatorial region can be estimated by eq. (1).

The total fallout of sulfuric acid (H_2SO_4) in a unit area at a pit site is obtained by multiplying the H_2SO_4 concentration of a layer, which includes non-sea-salt sulfate ion ($nssSO_4^{2-}$) originating from a volcanic eruption, by the water equivalent thickness of the layer. The $nssSO_4^{2-}$ concentration is calculated using the following equation, assuming that Na^+ ions are entirely derived from sea spray:

$$(SO_4^{2-})_{\text{non-sea-salt}} = (SO_4^{2-}) - (SO_4^{2-}/Na^+)_{\text{seawater}} \cdot (Na^+), \qquad (2)$$

where (SO_4^{2-}) and (Na^+) are the measured SO_4^{2-} and Na^+ concentrations of the snow sample, and $(SO_4^{2-}/Na^+)_{\text{seawater}}$ is the ratio of SO_4^{2-} to Na^+ concentrations of seawater being 0.25 in a weight ratio.

4. Estimate of SO_2 Emission from the 1982 Eruption of El Chichon Volcano

We show the concentrations of Na^+, SO_4^{2-} and $nssSO_4^{2-}$ in Table 1. Two significant $nssSO_4^{2-}$ signals were found in layers between 0.06 and 0.12 m depths and between 0.43 and 0.45 m depths, which correspond to 1993 and 1983, respectively (Fig. 2b). Judging from its date, the deeper signal is likely due to the deposition of sulfuric acid aerosols from the 1982 eruption of El Chichon volcano (in Mexico, 17°36′N, 93°23′W). In 1982, El Chichon volcano emitted 7 Mt of SO_2 to the stratosphere, high enough to give a clear $nssSO_4^{2-}$ signal in polar regions (BLUTH *et al.*, 1993; KOHNO *et al.*, 1996). The $nssSO_4^{2-}$ signal found between 0.06 and 0.12 m depths may include sulfuric acid aerosols originating from the 1991 eruption of Pinatubo volcano (in the Philippines, 15°14′N, 120°35′E) and the 1991 eruption of Cerro Hudson (in Chile, 46°17′S, 72°92′W). BLUTH *et al.* (1993) reported the SO_2 emission measured by TOMS during the eruption of Pinatubo volcano to be 20 Mt SO_2 and that of Cerro Hudson volcano to be 2 Mt SO_2. The Cerro Hudson volcano is closer to Antarctica and thereby yielded substantial amounts of $nssSO_4^{2-}$ at the pit site. Since the signal cannot be easily decomposed into the Pinatubo and Cerro Hudson components, we discuss only the emission from the 1982 eruption of El Chichon here.

The deposition of $nssSO_4^{2-}$ in the layer between 0.43 and 0.45 m depths, corresponding to 1983, is 4.2 $kg \cdot km^{-2}$. The deposition of $nssSO_4^{2-}$, due to the eruption of El Chichon, was obtained as 2.1 $kg \cdot km^{-2}$ by subtracting the background $nssSO_4^{2-}$ (2.1 $kg \cdot km^{-2}$) originating from oxidation of dimethylsulfide (DMS) produced by ocean biogenic activity. The background $nssSO_4^{2-}$ level was obtained by averaging all layers

Table 1. Chemical composition of snow from the surface to 1.9 m depth in a snow pit at Dome Fuji Station.

Water equivalent in m		Na^+	SO_4^{2-}	$nssSO_4^{2-}$
Upper depth	Lower depth	ng · g^{-1}	ng · g^{-1}	ng · g^{-1}
0.00	0.01	1.8	32.8	32.3
0.01	0.02	4.6	61.1	59.9
0.02	0.04	11.4	85.9	83.1
0.04	0.05	8.3	86.7	84.6
0.05	0.06	142.2	127.6	124.0
0.06	0.08	21.9	204.1	198.6
0.08	0.11	21.3	231.1	225.7
0.11	0.12	43.3	247.5	236.6
0.12	0.13	3.8	161.7	160.8
0.13	0.14	2.1	108.0	107.5
0.14	0.15	32.2	125.9	117.8
0.15	0.17	27.5	123.8	116.9
0.17	0.19	14.2	96.6	93.1
0.19	0.22	42.5	141.4	130.8
0.22	0.23	3.2	79.0	78.1
0.23	0.26	15.0	124.7	121.0
0.26	0.27	32.7	112.7	104.5
0.27	0.29	1.1	41.5	41.3
0.29	0.30	1.9	41.5	41.0
0.30	0.31	8.9	95.9	93.7
0.31	0.33	16.6	109.7	105.5
0.33	0.34	15.1	108.0	104.2
0.34	0.36	15.0	109.1	105.3
0.36	0.37	40.5	172.0	161.9
0.37	0.39	23.7	117.7	111.8
0.39	0.42	15.9	98.2	94.2
0.42	0.43	30.1	113.3	105.8
0.43	0.45	32.4	116.7	108.6
0.45	0.48	11.4	217.1	214.2
0.48	0.49	20.8	139.1	133.9
0.49	0.50	1.8	63.4	63.0
0.50	0.51	18.5	80.7	76.1
0.51	0.53	19.3	102.1	97.3
0.53	0.55	33.6	97.3	88.9
0.55	0.57	26.9	88.5	81.8
0.57	0.59	24.5	105.4	99.2

except the volcanic layers (Table 2 and Fig. 2b).

Deposition of the total β activity, derived from nuclear tests in 1952 and 1954, at DF-80 near Dome Fuji Station was found to be 2.84×10^{-1} mCi·km^{-2} by integrating the total β activity in the layers between 0.83 and 1.04 m in the snow pit in 1985. These

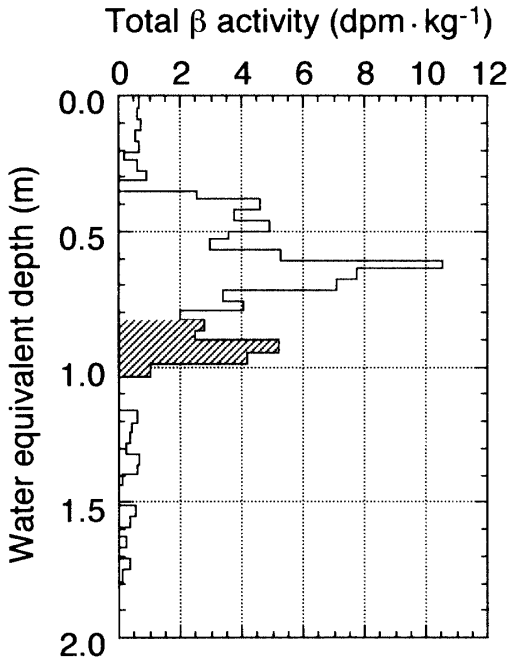

Fig. 3. Vertical profile of total β activity of snow pit in December 1985 at DF-80 near Dome Fuji Station (KAMIYAMA et al., 1997). Unit of dpm means disintegration per minute (1 dpm = 1/ 3.7×10^{-7} mCi). A hatched area indicates a layer of total β activity produced by the nuclear tests in 1952 and 1954.

depths correspond to the middle of 1953 and the end of 1959, respectively (Fig. 3; data after KAMIYAMA *et al.*, 1997). These dates were determined by considering a time difference of ~3 years (POURCHET and PINGLOT, 1979) between nuclear tests and appearance of the total β activity peak on Antarctica, and their residence time in the stratosphere of ~1.5 years (POURCHET *et al.*, 1983). The $\beta^{\text{pit site}}$ value in eq. (1) was corrected for radioactive decay between the time of deposition and measurement under the assumption that the total β activity was due only to ^{90}Sr ($T_{1/2}=28$ years) and ^{137}Cs ($T_{1/2}=29$ years). The corrected $\beta^{\text{pit site}}$ is calculated to be 7.53×10^{-1} mCi·km^{-2} assuming that the total β activity was mainly due to ^{90}Sr. The $\beta^{\text{test region}}$ has been reported to be 9.56×10^{9} mCi as mentioned above (UNSCEAR Report, 1982; CLAUSEN and HAMMER, 1988). Thus, a scaling factor, the ratio of $\beta^{\text{test region}}$ to $\beta^{\text{pit site}}$, is calculated to be 1.3×10^{10} km^2 for the region around Dome Fuji Station (Table 2).

The amount of SO_2 emission from the 1982 eruption of El Chichon is thus

Table 2. Glaciological SO_2 estimate of the 1982 eruption of El Chichon.

SO_4 deposition			$\beta^{\text{test region}}/\beta^{\text{pit site}}$	SO_2^{GE}	SO_2^{SE}
Total kg·km^{-2}	Background kg·km^{-2}	Volcanic kg·km^{-2}	km^2	Mt	Mt
4.2	2.1	2.1	1.3×10^{10}	17	7

GE: glaciological estimate; SE: spectroscopic estimate (BLUTH *et al.*, 1993).
Note: $\beta^{\text{test region}}=9.56\times10^{9}$ mCi (CLAUSEN and HAMMER, 1988); $\beta^{\text{pit site}}=7.53\times10^{-1}$ mCi after correction for radioactive decay.

calculated to be 17 ± 1 Mt SO_2 using eq. (1). The error is propagated from the following factors. The error in layer thickness is considered to be within 0.5%. Analytical error of the SO_4^{2-} determination with ion chromatography has been reported to be within 0.3% (IGARASHI *et al.*, 1998). The error for the total β activity determination is estimated to be within 7% for the activity level of 3 dpm·km^{-2} (KAMIYAMA, pers. commun.), the mean total β activity emitted in 1952 and 1954. The error in the estimation of the amount of total β activity produced by bomb-tests has not been reported.

The glaciological estimate of the SO_2 emission of 17 ± 1 Mt SO_2 is apparently greater than the spectroscopic estimate of 7 Mt SO_2 (BLUTH *et al.*, 1993). According to SYMONDS *et al.* (1994), the spectroscopic estimate which was directly measured in the volcanic cloud by a satellite-carried TOMS is considered to be an underestimate because low SO_2 concentration at the margin of the volcanic cloud is below the detection limit of TOMS measurement.

While much work remains to be done to determine the reason for the difference between glaciological and spectroscopic estimates (17 and 7 Mt SO_2, respectively), the ratio between glaciological and spectroscopic estimates (~2.5) is much less than the ratio (~100) between spectroscopic and petrologic estimates (7 and 0.07 Mt SO_2, respectively; BLUTH *et al.*, 1993; DEVINE *et al.*, 1984) by a factor of 40. This gives us to a hope that glaciological estimation will be a useful tool for paleoclimatological studies. Thus, it may be said that SO_2 emission by past explosive volcanic eruptions in the equatorial region can be estimated by the glaciological method, and that assessment of the impact of past explosive volcanic eruptions on the coeval global climate may be made possible through the study of volcanic signals in glaciers and ice sheets.

5. Conclusions

The SO_2 emission from the 1982 eruption of El Chichon was estimated to be 17 ± 1 Mt SO_2 by the glaciologocal method using nssSO_4^{2-} concentration in snow samples collected near Dome Fuji Station, Antarctica. This glaciological estimate of SO_2 emission is consistent with the reported spectroscopic estimate of 7 Mt SO_2. This suggests that the glaciologocal method is applicable to estimate SO_2 emission from past explosive volcanic eruptions.

Acknowledgments

We acknowledge Ms. T. KOBAYASHI and Dr. M. IGARASHI of the National Institute of Polar Research for their help in the chemical analysis. We thank Dr. T. FUKUOKA of Rissho University and Mr. N. HAYASHI of Nihon University for their help in the sample preparation. We appreciate the help of the 37th and 38th Japanese Antarctic Research Expeditions, during which snow near Dome Fuji Station was collected.

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(Received May 1, 1998; Revised manuscript accepted June 5, 1998)